



03-07-06

AF\$

PATENT

gk  
Ifw

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Hoke, et al.

)

)

)

)

)

)

)

)

Examiner: Hien Thi Tran

Serial No.: 10/002,272

Group Art Unit: 1764

Filed: November 15, 2001

Docket: 4569 D DIV

For: Method and Device for Cleaning the Atmosphere

**COVER LETTER ENCLOSING BRIEF ON APPEAL**

Mail Stop Appeal Brief - Patents

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450

Sir:

The Appeal Brief is presented in support of the Notice of Appeal to the Board of Patent Appeals and Interferences, filed on December 6, 2005, from the Final Rejection of claims 48-53 and 55-58 of the above-identified application, as set forth in the Final Office Action mailed on June 10, 2005.

This Appeal Brief is filed in triplicate. A Credit Card Payment Form is enclosed in payment of the \$500.00 large entity fee. A Petition for a one-month extension of time is enclosed. The Commissioner of Patents is hereby authorized to charge any underpayment or credit any overpayment to Deposit Account No. 50-3329. The Appellants respectfully request consideration and reversal of the Examiner's rejections of pending claims.

The undersigned was authorized by Richard A. Negin, Reg. No. 28,649, an attorney of record in the subject application, to prepare and file this Appeal Brief on

behalf of the Assignee. Correspondence should continue to be directed to Chief Patent Counsel,  
Engelhard Corporation, 101 Wood Avenue, P.O. Box 770, Iselin, NJ, 08830-0770.

Respectfully submitted,

Date: March 6, 2006

By SSSsla

Scott S. Servilla

Reg. No. 40806

Telephone (732) 815-0404

Engelhard Corporation  
101 Wood Avenue  
P.O. Box 770  
Iselin, NJ, 08830-0770

CERTIFICATE UNDER 37 CFR 1.10: The undersigned hereby certifies that this correspondence is being deposited with the United States Postal Service with sufficient postage as Express Mail (EQ 182 836 740 US) in an envelope addressed to: Mail Stop Appeal Brief, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on March 6, 2006.

SSSsla

Signature

Name: Scott S. Servilla



Docket No.: 4569D (DIV)  
(PATENT)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:  
Hoke et al.

Application No.: 10/002,272

Filed: November 15, 2001

For: METHOD AND DEVICE FOR CLEANING  
THE ATMOSPHERE

:  
:  
:  
: Group Art Unit: 1764  
: Examiner: H.T. Tran  
:  
:  
:  
:  
:  
:  
:

Mail Stop Appeal Brief-Patents  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

APPELLANT'S BRIEF

03/08/2006 DEMMANU1 00000006 10002272

01 FC:1402

500.00 OP

I hereby certify that this correspondence is being deposited with the U.S. Postal Service with sufficient postage as Express Mail (EQ 182 836 740 US), in an envelope addressed to: Mail Stop Appeal Brief-Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the date shown below.

Dated: March 6, 2006

Signature

Scott S. Servilla

## **TABLE OF CONTENTS**

	<b><u>Page</u></b>
Real Party in Interest.....	3
Related Appeals and Interferences.....	3
Status of Claims.....	3
Status of Amendments.....	3
Summary of Claimed Subject Matter.....	3
Grounds of Rejection to be Reviewed on Appeal.....	5
Argument.....	5
Conclusion.....	14
Claims Appendix.....	15
Evidence Appendix.....	17

### **Real Party in Interest**

The real party in interest in this proceeding is Engelhard Corporation, the Assignee of record.

### **Related Appeals and Interferences**

Neither Appellant nor its agents are aware of any prior or pending appeals, judicial proceedings or interferences which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

### **Status of Claims**

Claims 1-36 have been cancelled, and claims 37-47 have been withdrawn. Claims 48-53 and 55-58 stand finally rejected under 35 U.S.C. § 103(a) and are appealed. A copy of the claims on appeal are in the Claim Appendix of this Brief.

### **Status of Amendments**

No amendments have been submitted subsequent to the Final Rejection mailed on June 10, 2005.

### **Summary of Claimed Subject Matter**

Claim 48 is directed to a device for treating the atmosphere to catalytically convert atmospheric pollutants to less harmful materials comprising an outer surface of a motor vehicle component 100 which is exposed to a flow of ambient air, the outer surface being coated with a catalyst composition 102 that is protected with a porous overcoat of carbon 104. (Page 4, line 19 through page 5, line 25; Fig. 1A.)<sup>1</sup>

Claim 49 depends from claim 48, and further requires one hydrophobic protective material 104 overcoating the overcoat of carbon, the protective material capable of

---

<sup>1</sup> Page and paragraph designations refer to those found in the originally filed specification.

substantially preventing liquid water and/or water vapor from reaching the catalytic composition or overcoat of carbon. (Page 6, lines 21-24; Fig. 1A.)

Claim 50 depends from claim 48, and further requires at least one layer of the hydrophobic material overcoating the catalyst composition or overcoat of carbon. (Page 6, lines 6-7.) Claim 51 depends from claim 48 and further requires that the catalyst composition is selected from base metals, precious metals, salts and oxides thereof and combinations thereof. (Page 13, line 21 through page 14, line 7.) Claim 52 depends from claim 48 and further requires that the catalyst comprises manganese dioxide. (Page 13, lines 23-24.) Claim 53, depending from claim 48, requires that the overcoat of carbon is selected from activated granular carbon, carbon black, permanganate on carbon, and mixtures thereof. (Page 12, lines 24-26.) Claim 55 depends from claim 49 and requires that the hydrophobic material is selected from fluoropolymers and silicone polymers. (Page 13, lines 4-19.)

Claim 56 depends from claim 48 and recites that the outer surface of the motor vehicle component is selected from the outer surface of radiators, air-conditioner condensers, charge air coolers, transmission coolers and inserted devices. (Page 5, lines 20-24.) Claim 57 depends from claim 57 and specifies that the catalyst composition is selected from manganese dioxide, platinum, palladium and mixtures thereof. (Page 6, lines 25-28.) Claim 58 depends from claim 48 and specifies that the catalyst composition is supported on a material selected from ceria, alumina, titania, silica, zirconia and mixtures thereof. (Page 14, lines 9-13.)

### **Grounds of Rejection to be Reviewed on Appeal**

1. Whether claims 48-53 and 55-58 are unpatentable under 35 U.S.C. § 112, second paragraph, as being indefinite.
2. Whether claims 48-53 and 56-58 are unpatentable under 35 U.S.C. § 103(a) over WO 98/022235 in view of JP 52-122290.
3. Whether claim 55 is unpatentable under 35 U.S.C. § 103(a) over WO 98/022235 in view of JP 52-122290 further in view of Hoke et al. United States Patent No. 6,190,627.

### **Argument**

Claims 48-53 and 55-58 in this application stand finally rejected under 35 U.S.C. § 112, second paragraph, as allegedly being indefinite. Claims 48-53 and 56-58 in this application stand finally under 35 U.S.C. § 103(a) as allegedly unpatentable over WO 98/022235 in view of JP 52-122290. Claim 55 in this application stand finally rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over WO 98/022235 in view of JP 52-122290 further in view of Hoke et al. United States Patent No. 6,190,627. Claims 48-53 and 55-58 in this application stand rejected under the judicially created doctrine of obviousness-type double patenting as allegedly unpatentable over claims 6,190,627 in view of JP 52-122290.<sup>2</sup>

### **Rejection of Claims 48-53 and 55-58 Under 35 U.S.C. § 112**

In the final office action, the Examiner rejected the claims, stating “[i]n claim 48, line 3 it is unclear as to how the ambient air is related to the atmosphere set forth in line 1.” (Final Office Action, Page 2, paragraph 2.)

---

<sup>2</sup> In an amendment submitted March 14, 2005, applicants acknowledged the obviousness-type double patenting rejection and stated that a terminal disclaimer would be filed upon allowance of at least one pending claim.

The Manual of Patent Examining Procedure requires that “the words of the claim must be given their plain meaning unless applicant has provided a clear definition in the specification.” (MPEP Section 2111.01 (citations omitted.)) Accordingly, it is fundamental under 35 U.S.C. 112, second paragraph that applicants can be their own lexicographers and that they “can define in the claims what they regard as their invention essentially in whatever terms they choose so long as any special meaning assigned to a term is clearly set forth in the specification.” (MPEP Section 2173.01.) The MPEP cautions that examiners “should allow claims which define the patentable subject matter with a reasonable degree of particularity and distinctness” and that “[s]ome latitude in the manner of expression and the aptness of terms should be permitted even though the claim language is not as precise as the examiner might desire.” (MPEP 2173.02.)

In the amendment submitted on March 14, 2005, applicants pointed out that the specification clearly establishes the relationship between the terms “atmosphere” and “ambient air” in the application. In the specification, “ambient air” is defined as “the portion of the atmosphere that drawn or forced toward the outer surface of the coated substrate” (see page 5, lines 12-13 of the specification). The specification defines “atmosphere” as “mass of air surrounding the earth” (see page 5, lines 10-11 of the specification).

Applicants respectfully submit that in view of the definitions in the specification, the relationship between “atmosphere” and “ambient air” is clear. Since the language of the claim is such that a person of ordinary skill in the art could interpret the metes and bounds of the claim so as to understand how to avoid infringement, the rejection of the



claim under 35 U.S.C. 112, second paragraph, is inappropriate. See MPEP Section 2173.02.

**Rejection of Claims 48-53 and 56-58 Under 35 U.S.C. § 103(a)**

Claims 48-53 and 56-58 were rejected in the Final Office Action as allegedly unpatentable under 35 U.S.C. § 103(a) over WO 98/02235 in view of JP-52-122290.<sup>3</sup> At page 3 of the Final Office action, WO 98/02235 is cited as disclosing a motor vehicle component, which is exposed to a flow of ambient air, the radiator being coated with a catalyst material selected from a base metal, a precious metal or manganese oxide and a protective layer of polymers. In rejecting the claims, the Examiner states that “WO 98/02235 recognizes that the ambient air contains pollutants such as CO.” (Final Office Action, at page 3, paragraph 5.) The Examiner relies upon JP52-122290 for the teaching of “the conventionality of providing a porous layer of materials, such as active carbon to cover the catalyst surface to prevent catalyst poisoning.” (Final Office Action, Page 3, paragraph 5.) The Examiner concludes that it would have been obvious to provide a porous overcoat of carbon material on the surface of the catalyst material as taught by JP 52-122290 in addition to the porous protective layer in the apparatus of WO 98/02235 to prevent further catalyst poisoning. (Id.)

To establish a prima facie case of obviousness, three basic criteria must be met. There must be some suggestion or motivation from the references themselves or the knowledge generally available to one of ordinary skill in the art to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. Third, the teaching or suggestion to make the claimed combination and the

---

<sup>3</sup> The Office Action cited JP 52-122290, but only the Abstract for this document was provided. Under MPEP Sections 1207.02 and 607.02, a copy of a translation of JP 52-122290 is submitted with this Appeal Brief.

reasonable expectation of success must both be in the prior art, and not on the applicants' disclosure. (MPEP Section 2142.)

In addition, in determining the differences between the prior art and the claims, the question is not whether the differences themselves would have been obvious, but whether the claimed invention as a whole would have been obvious. MPEP Section 2141.02. Subheading III of MPEP Section 2141.02 notes that discovering the source/cause of a problem is part of the "as a whole" inquiry. This subheading quotes *In re Spinnoble*, 405 F.2d 578, 585, 160 USPQ 237, 243 (CCPA 1969), the pertinent part stating that "a patentable invention may lie in the discovery of the source of a problem even though the remedy may be obvious once the source of the problem is identified. This is part of the 'subject matter as a whole' which should always be considered in determining the obviousness of an invention under 35 U.S.C. § 103."

Continuing the discussion of *In re Spinnoble*, MPEP section 2141.02 states:

In *In re Spinnoble*, the claim was directed to a plural compartment mixing vial wherein a center seal plug was placed between two compartments for temporarily isolating a liquid-containing compartment from a solids-containing compartment. The claim differed from the prior art in the selection of butyl rubber with a silicone coating as the plug material instead of natural rubber. The prior art recognized that leakage from the liquid to the solids compartment was a problem, and considered the problem to be a result of moisture passing around the center plug because of microscopic fissures inherently present in molded or blown glass. The court found the inventor discovered the cause of moisture transmission was through the center plug, and there was no teaching in the prior art which would suggest the necessity of selecting applicant's plug material which was more impervious to liquids than the natural rubber plug of the prior art.

Furthermore, in concluding that the claimed invention was patentable over the prior art, the *Sponnoble* court noted that a reference to Parsons, which mentioned moisture permeability, did not appear to be directed to permeability of liquid water, stating "[t]hat a natural rubber plug might be permeable to steam in no way establishes or makes obvious its permeability to liquid water; thus the cause of the problem is not suggested by the prior art." 405 F.2d at 586-87.

In the instant case, like in *Sponnoble*, Applicants respectfully submit that the Examiner has failed to establish *prima facie* obviousness. Applicants recognize that WO 98/02235 teaches a catalyzed surface of a motor vehicle component to convert atmospheric pollutants such as CO to less harmful materials. However, WO 98/02235 teaches the use of a resistant, repellent overcoat to protect the underlying catalyst from roadway contaminants such as chemicals, salts, dirt and other contaminants which may adversely affect the catalyst composition. (WO 98/02235, Page 15, lines 25-33.) The Examiner also refers to WO 98/02235 as teaching that ambient air contains pollutants such as CO.

JP 52-122290 teaches the use of an adsorptive overcoat material to protect an exhaust gas catalyst used to purify a waste gas stream from automobiles, combustion furnaces, oil burners, incinerators and chemical plants to protect the catalyst from waste gas stream contaminants such as S, P, Pb, Ca, and Ni. The Examiner states that it would have been obvious to place the porous carbon overcoat of JP 52-122290 on the surface of the catalyst material in WO 98/02235 containing the resistive porous protective layer so as to prevent further catalyst poisoning.

In the invention claimed in the instant application, a porous layer of carbon overcoats the catalyst to protect the catalyst from contaminants such as solid or vaporized particulates and corrosive compounds such as salts and oxides of nitrogen and sulfur, which can mask, foul and/or poison the catalyst by adsorbing the contaminants and maintains the contaminants out of contact from the catalyst. The porous carbon coating protects against contaminants including particulate matter, high molecular weight hydrocarbons, water borne salts, aerosols, and gases such as NO<sub>x</sub> and SO<sub>x</sub>. (Specification, Page 8, lines 24-30.)

Applicants respectfully submit that the skilled artisan reading WO 98/02235 together with JP 52-122290 would not be motivated to apply one of the adsorptive coatings disclosed in JP 52-122290 over the resistive or repellent coating disclosed in WO 98/02235. First, WO 98/02235 pertains to the field of treating the atmosphere by contacting the catalyst contained on the surface of an automobile with ambient air containing pollutants to convert the pollutants into harmless compounds. There is no teaching or suggestion in WO 98/02235 to provide an adsorptive coating over the resistive coating to further prevent catalyst poisoning. To do so would render the device taught in the reference unsuitable for its intended purpose, which is to repel contaminants from the catalyst surface. If the proposed modification of a reference renders the reference unsuitable for its intended purpose, there can be no suggestion to make the proposed modification. See MPEP Section 2143.01 Subheading V.

Furthermore, JP 52-12290 pertains to catalysts for the treatment of waste gases, from automobiles, combustion furnaces, oil burners, incinerators and chemical plants, not ambient air. The various coatings in JP 52-12290 are described as being useful for

protecting the catalyst from S, Pb, Ca, and Ni in a waste gas stream. Applicants submit that it cannot be assumed that a material suitable for treating a waste gas stream would be suitable as a protective overcoat for a catalyst used in the treatment of ambient air because the catalysts encounter different temperatures, pollutant mix/concentration and other use conditions. There is no teaching or suggestion in JP 52-12290 that a porous carbon coating would be suitable for adsorbing the types of contaminants encountered by the catalysts of the present invention. It appears that the Examiner has relied on improper hindsight in assuming that the carbon coating used in a waste gas stream for protecting the catalyst from different types of contaminants would be successful in protecting the catalyst in the instant invention. There is no teaching in JP 52-12290 would be useful, and thus there is no reasonable expectation of success that the coating in JP 52-12290 would protect the catalyst of the instantly claimed invention. Accordingly, the combination of WO 98/02235 and JP 52-12290 cannot render claim 38 obvious.

Regarding claims 49 and 50 there is no teaching or suggestion in either reference relied upon by the Examiner to apply at least one overcoat of a hydrophobic protective material over the overcoat of carbon. Claims 51, 52 and 56-58 are patentable for at least the reasons provided above with respect to claim 48.

Regarding claim 53, there is no teaching or suggestion in WO 98/02235 or JP 52-12290 of the particular types of carbons recited in claim 53.

Regarding claim 55, the Examiner has rejected this claim as being unpatentable over WO 98/02235 in view of JP 52-122290 further in view of United States Patent No. 6,190,627. For at least the reasons provided above with respect to claim 48, 49 and 50, applicants submit claim 55 is patentable over WO 98/02235 in view of JP 52-122290.

Regarding the Examiner's reliance on United States Patent No. 6,190,672, applicants submit that since the present application claims priority to the application which issued as United States Patent No. 6,190,627, the '627 patent is not prior art to the present application. Accordingly, the Examiner has failed to establish a *prima facie* case of obviousness.

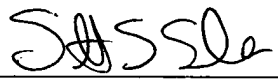
### **Conclusion**

In view of the foregoing, claims 48-53 and 55-58 are definite under 35 U.S.C. § 112. Furthermore, claims 48-53 and 56-58 of the application are not obvious under 35 U.S.C. § 103(a) over WO 98/02235 in view of JP 52-122290, and claim 55 is not obvious over WO 98/02235 in view of JP 52-122290 and further in view of United States Patent No. 6,190,627.

The undersigned was authorized by Richard A. Negin, Reg. No. 28,649, an attorney of record in the subject application, to prepare and file this Appellant's Brief on behalf of the Assignee. Correspondence should continue to be directed to Chief Patent Counsel, Engelhard Corporation, 101 Wood Avenue, P.O. Box 770, Iselin, NJ, 08830-0770.

Dated: March 6, 2006

Respectfully submitted,

By:   
Scott Servilla  
Registration No. 40,806  
Telephone: (732) 815-0404  
Facsimile: (732) 815-1330

Engelhard Corporation  
101 Wood Avenue  
P.O. Box 770  
Iselin, NJ, 08830-0770

### **Claims Appendix**

48. A device for treating the atmosphere to catalytically convert atmospheric pollutants to less harmful materials comprising an outer surface of a motor vehicle component which is exposed to a flow of ambient air, said outer surface being coated with a catalyst composition, said catalyst composition being protected with a porous overcoat of carbon.

49. The device of claim 48 further comprising at least one hydrophobic protective material overcoating the overcoat of carbon, said protective material being capable of substantially preventing liquid water and/or water vapor from reaching the catalyst composition or overcoat of carbon.

50. The device of claim 48 comprising at least one layer of the hydrophobic protective material coated over the catalyst composition or overcoat of carbon.

51. The device of claim 48 wherein the catalyst composition is selected from the group comprising base metals, precious metals as well as salts and oxides thereof and combinations thereof.

52. The device of claim 48 wherein the catalyst composition comprises manganese dioxide.

53. The device of claim 48 wherein the overcoat of carbon is selected from the group comprising activated granular carbon, carbon black, permanganate on carbon and mixtures thereof.

55. The device of claim 49 wherein the hydrophobic protective material is selected from the group comprising fluoropolymers and silicone polymers.

56. The device of claim 48 wherein said outer surface of a motor vehicle component is selected from the outer surface of radiators, air-conditioner condensers, charge air coolers, transmission coolers and inserted devices.

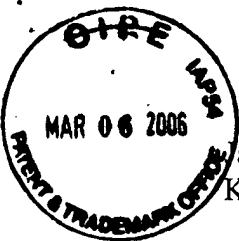
57. The device of claim 48 wherein said catalyst composition is selected from manganese dioxide, platinum, palladium and mixtures thereof.

58. The device of claim 48 wherein the catalyst composition is supported on a material selected from ceria, alumina, titania, silica, zirconia, and mixtures thereof.



### **Evidence Appendix**

Translation of JP 52-122290, cited in Final Office Action at Page 2 and Office  
Action mailed on October 14, 2004 at page 3.



Japan Patent Office

Kokai Patent Publication No. Sho 52 (1977) – 122290, October 14, 1977

Catalyst for use in the treatment of exhaust gas

Patent Application No. Sho 51 (1976) – 39069

Date of Application: April 7, 1976

Inventor: S. Adachi

c/o Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo

Inventor: A. Koyama

c/o Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo

Applicant: Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo

---

Continued to the last page

---

### Specification

#### (1). Title of Invention

Catalyst for use in the treatment of exhaust gas

#### (2). Claims of the Patent

[1]. Catalyst for use in the treatment of exhaust gas, the catalyst being used at the treatment of exhaust gas and characterized by that at least a part of the surface of the catalyst is covered with the layer of a porous material that is different from the catalyst component.

[2] The catalyst for use in the treatment of exhaust gas described in Claim 1 in which the material that covers at least a part of the surface of the catalyst is at least one that is selected from aluminum oxide, active carbon, alkali metal oxide, alkali earth metal oxide, the metal oxide or sulfate of the transition metal such as iron, manganese, copper, nickel, cobalt, zinc, cadmium, etc.

#### (3). Detailed Description of the Invention

This invention is related to the catalyst for use in the treatment of exhaust gas that contains nitrogen oxide. More specifically, this invention is related to the catalyst which is suitable in the treatment of exhaust gas for the effective reduction and removal of NO<sub>x</sub> in the gas that simultaneously contains nitrogen oxide (hereinafter, this is called NO<sub>x</sub> and sulfur oxide.

The pollution of atmospheric air by the NO<sub>x</sub> which is contained in the exhaust gas from automobiles and in the waste gas from the various combustion furnaces, heavy oil burners, incinerators, chemical plants, etc. are being discussed as a social problem and the appearance of an effective means of making it harmless is being desired.

Up to the present, as for the means of removing the NO<sub>x</sub> contained in the waste gas, the method of passing it to a washing solution to have it absorbed, the method of having it adsorbed to an adsorbing agent, the method of having it contact with a catalyst to convert it to a harmless material, etc. were known.

However, the washing method uses a large amount of liquid and so the handling is inconvenient and also it requires facilities of a large scale and the adsorbing method is unsuitable for the treatment of a large amount of the waste gas. For this reason, the method of treatment by using a catalyst has been drawing attention and, as the catalyst for use in this treatment of waste gas, the metals of platinum family, the oxides of metals such as iron, manganese, copper, molybdenum, vanadium, etc., or the catalyst prepared by having these carried in a carrier such as alumina, silica, etc. were proposed. However, the catalysts said above which have been known from the past are weak against the poisoning primarily by the sulfur oxide contained in the waste gas and so they have had problems in terms of the life.

The present invention eliminates the shortcomings described above and provides an inexpensive catalyst in which the catalyst component is not poisoned by the compounds of sulfur, lead, calcium, nickel and the steam, vanadium compound, nickel compound, etc in the waste gas containing nitrogen oxide and so the catalyst activity is maintained stably for a long time.

Thus, this invention is a catalyst that is used for converting harmless the nitrogen oxide in the waste gas and, by making a double structure in which a part of the catalyst surface is covered with a porous material that is different from the catalyst being used and by this, the compounds of sulfur, phosphorus, lead, etc are adsorbed selectively to the porous material to prevent the adsorption or sticking to the catalyst component and prevent the degradation of the catalyst activity. Further, the severe degradation of the strength of catalyst is prevented.

As for the porous material which is used in this invention, the examples are: aluminum oxide, active carbon or the alkali metal oxide such as Na<sub>2</sub>O, K<sub>2</sub>O, etc.; the alkali earth oxide such as CaO, SrO; the oxides of transition metal such as Fe, Mn, Cu, Ni, Co, Zn, Cd, etc., their mixture and compound; also their compounds such as sulfate.

Also, the porous material that is different from the catalyst is for protecting the catalyst by covering the catalyst surface with a material that does not necessarily have the catalyst activity based on the key point of this invention and it means to have a proper porosity so that it does not give a large hindrance to the movement of the reactant and product to and from the catalyst surface. As to the thickness of its covering, 5 μm ~ 1000 μm is sufficient and, as for the covering ratio, 10 % ~ 100 % relative to the catalyst surface area is

sufficient. Further, as for the catalyst component which is used in this invention, there is no particular restriction as long as it is an effective one as the catalyst for use in the treatment of waste gas, including those that are known as the catalyst for use in the treatment of waste gas such as the platinum- carrying catalyst, the sintered oxide of transition metal such as vanadium, iron, copper, nickel, cobalt, manganese, chromium, molybdenum, tungsten, etc and those in which these are carried on the carriers and those in which their sulfate is carried on carrier. This is natural in view of the key point of this invention.

As for the waste gas that is the object of treatment of this invention, there are the boiler waste gas, and the waste gas from various types of combustion furnaces, heavy oil burners, incinerators, chemical plants, automobiles, etc. containing NO<sub>x</sub>,

As for the reducing gas that is added when necessary in the catalyzed reduction of NO<sub>x</sub>, there are the carbon monoxide, hydrogen sulfide, hydrogen, hydrocarbon or their mixture, or the water gas, etc.

In the following examples of application, ammonia is used as the reducing gas but, as is clear from the key point of this invention, it is not limited to this and, also, it goes without saying that the catalyst that is used is not limited to these examples of application.

By using the catalyst of this invention, for example, when ammonia is added as the reducing gas into the waste gas containing NO<sub>x</sub> and, in the temperature range of 200 ~ 600 deg C, the treatment is conducted at the spatial velocity of  $1 \times 10^3 \sim 1 \times 10^5 \text{ hour}^{-1}$  (translator's note: the power indices of 10, i.e. 3 and 5 here are not clearly legible in the original copy of the patent; check with the more clearly legible copy), conversion of the waste gas to harmless gas can be conducted for a long time with a high efficiency of cleaning. The amount of addition of the ammonia needs to be at least at 2/ 3 mol ratio with respect to the nitrogen oxide but, when a complete cleaning is not necessarily required, the amount of ammonia can be reduced.

In the following, detailed explanation is given by using the examples of application.

#### Example of Application 1

In the carrier made of porous alumina of spherical diameter of 6 mm, by using the ordinary method of impregnation, iron oxide and vanadium oxide were carried by a ratio of Fe/ V = 7/ 3 (atomic ratio). It was confirmed that the component of the carried substance was contained by 5.0 wt % with respect to the alumina. (This catalyst is designated as A).

On the surface of the catalyst A, by using the ordinary pan type granulating machine, Fe<sub>2</sub>O<sub>3</sub> was applied and baking was done at 500 deg C to for the covering (This catalyst is designated as B.). Under a microscope observation, it was confirmed to have a double structure in which the Fe<sub>2</sub>O<sub>3</sub> layer of about 50 μm was covering the surface.

The catalysts A, B that were obtained as described above were filled in a catalyst tank and this was maintained at 300 deg C and, to this, the waste gas from a boiler which was

burning the heavy oil B (NO approximately 100 ppm, SO<sub>2</sub> 150 ppm, CO<sub>2</sub> 10 %, steam 10 %, O<sub>2</sub> 4 %, remainder N<sub>2</sub>) was passed at a spatial velocity of 5000 hr<sup>-1</sup> with respect to the amount of the catalyst for 1000 hours. (These catalysts are designated as A', B', respectively).

With these catalysts A, A', B, B' and the catalyst B'' obtained by removing the Fe<sub>2</sub>O<sub>3</sub> layer of the surface of the catalysts B', 40 cc of each catalyst was filled in a reactor tube made of quartz glass and the mixed gas consisting of NO 150 ppm, SO<sub>2</sub> 150 ppm, NH<sub>3</sub> 150 ppm, O<sub>2</sub> 1.5 %, steam 10 %, and remainder of nitrogen was passed at a spatial velocity of  $5 \times 10^3$  hr<sup>-1</sup> and the NO cleaning ratio,  $[(\text{NO (inlet)} - \text{NO (outlet)}) / \text{NO (inlet)}] \times 100$ , was measured. Also, the elements at the surface of these catalysts were analyzed by the fluorescent X ray analysis. From the sample after a 1000 hours test, sulfur and a small amount of phosphorus (translator's note: the phosphorus here was not clearly legible in the patent copy and it may be other substance) were detected in addition to the catalyst component. Among these, the result of measurements for the sulfur and the cleaning ratio are shown in Table 1.

Table 1

	Cleaning ratio at 300 deg C	S (K or primary)*
A	80 %	-
A'	75 %	600 cps
B	78 %	-
B'	77 %	650 cps
B''	79 %	30 cps

\* Translator's note: The word "primary" could be read as "first order" or "first time" but exactly what it means is not clear from the context. What the "cps" stands for is not clear also.

#### Example of Application 2

By the same method as in Example of Application 1, to the surface of catalyst A of Example of Application 1, Mn<sub>2</sub>O<sub>3</sub> layer of about 70 μm was covered to obtain the catalyst C. This catalyst C was tested under the same condition as in Example of Application 1 and the result is shown in Table 2. Also, as in Example of Application 1, the catalyst C' is the catalyst C that was exposed to the boiler waste gas for 1000 hours and the catalyst C'' is the catalyst C' from whose surface the Mn<sub>2</sub>O<sub>3</sub> was removed.

Table 2

	Cleaning ratio at 300 deg C	S (K or primary)*
A	80 %	-
A'	75 %	600 cps
C	77 %	-
C'	76 %	680 cps
C''	79 %	10 cps

\* Translator's note: The word "primary" could be read as "first order" or "first time" but exactly what it means is not clear from the context. What the "cps" stands for is not

clear also.

As is clear from these results, with A, A', the active components received the poisoning of the sulfur oxide, etc. in the waste gas component and so the activity degraded while, with the catalyst B, B', B'', C, C', C'', the component of sulfur oxide, etc. (believed to be mainly SO<sub>2</sub>) was substantially adsorbed to the Fe<sub>2</sub>O<sub>3</sub>, Mn<sub>2</sub>O<sub>3</sub> that covered the surface and so it did not reach the catalyst component and thus it is seen that there was less degradation of the catalyst activity. Also, when other catalyst component was used as in Example of Application 1 and the test was conducted on the catalyst of this invention that had the double structure covered with various porous materials which were different from the catalyst component, good results were obtained similarly.

Continuation of p. 1

Inventor: M. Takayama

c/o Tokyo Denki Kagaku Kogyo (Tokyo Electrochemical Industries) K. K.  
14- 6 2- Chome, Uchi Kanda, Chiyoda Ku, Tokyo